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Compression/Injection Molding of Bipolar Plates for Proton Exchange Membrane Fuel Cells

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Compression/Injection Molding of Bipolar Plates for Proton Exchange Membrane Fuel Cells

by

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Thesis

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To my mother, *for making me believe in myself*

To my father, *for teaching me the art of thinking big*

To my teachers, *for all the knowledge instilled in me*

To Perumal*, for "Mam Ekam Saranam Vrajah"*

and of course

To my little brother, *for putting up with me all the time*

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Abstract

Compression/Injection Molding of Bipolar Plates for Proton Exchange Membrane Fuel Cells

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Fuel cells are electrochemical energy conversion devices that convert chemical energy to electrical energy efficiently. Bipolar plates form an integral part of a fuel cell and their high manufacturing cost and low production rate have hindered the commercialization of fuel cells. Bipolar plates require high electrical conductivity, strength, chemical resistance and thermal conductivity. This thesis presents efforts to manufacture bipolar plates which meet these requirements using compression or injection molding. Compression or injection molding processes allow cost-effective, large-scale manufacturing of bipolar plates. A variety of material systems for the fabrication of bipolar plates are processed, molded and characterized.

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Chapter 1: *Introduction*

FUEL CELL

Fuel cells are promising energy conversion devices (Joon, 1998) for various portable electronic devices, stationary and mobile applications that operate at low temperatures (Shen, 2006). Fuel cells are electrochemical devices that have the capability to convert chemical energy in fuel directly into electrical energy.

The electricity in a fuel cell is generated through an electrochemical reaction which is initiated by a catalyst between the fuel on the anode side and the oxidant on the cathode side. The fuel cell is a thermodynamically open system and can theoretically produce energy as long as it is supplied with the fuel and oxidant. Since the energy conversion from the fuel to electricity is done in a single step it is more efficient, clean and reliable than fossil fuel combustion systems (Hayre, 2006). The most common type of fuel cells and their characteristics are shown in Table 1 (Larminie, 2000).

Table 1: Summary of common fuel cell types and characteristics

	Electrolyte ion	Cell efficiency	Operating temperature	Applications	Fuel
Proton Exchange Membrane Fuel Cell (PEMFC)	H^+	30–70%	20–80°C	Portable electronic devices, stationary and mobile applications	Hydrogen, methanol
Alkaline Fuel Cell (AFC)	H^+	50–70%	50–200°C	Space vehicles	Hydrogen
Phosphoric Acid Fuel Cell (PAFC)	OH^-	55%	~200°C	Small size stationary power systems	Natural gas

Molten Carbonate Fuel Cell (MCFC)	CO_3^{2-}	55%	~650°C	Medium to large size stationary power systems	Natural gas, biogas
Solid Oxide Fuel Cell (SOFC)	O^{2-}	60–65%	600–1000°C	Stationary power systems	Methane, butane, propane

Table 1: continued

PROTON EXCHANGE MEMBRANE FUEL CELL (PEMFC)

Low operating temperature and high power density make proton exchange membrane fuel cell (also known as Solid Polymer Fuel Cells - SPFC) a promising choice for various electronic and mobile applications. Direct methanol fuel cell (DMFC) is one of the most widely researched PEMFC systems. DMFC employs methanol as the fuel and is preferred over the other variants of PEMFC for portable devices. This is because methanol is easier to store and transport when compared to the other PEMFC fuels such as hydrogen. A typical DMFC single cell consists of a Membrane Electrode Assembly (MEA) sandwiched between two current collectors. The MEA consists of a proton exchange membrane (PEM) sandwiched between an anode and a cathode. The PEM electrically insulates the anode and the cathode and is permeable to protons (Barbir, 2005). The schematic of a DMFC is shown in Figure 1. The half cell electrochemical reactions are written as follows:

Anode oxidation of methanol: $CH_3OH + H_2O \rightarrow CO_2 + 6H^+ + 6e^-$

Cathode reduction of oxygen: $1½O_2 + 6H^+ + 6e^- \rightarrow 3H_2O$

Total DMFC reaction: $CH_3OH + 1½O_2 \rightarrow CO_2 + 2H_2O$

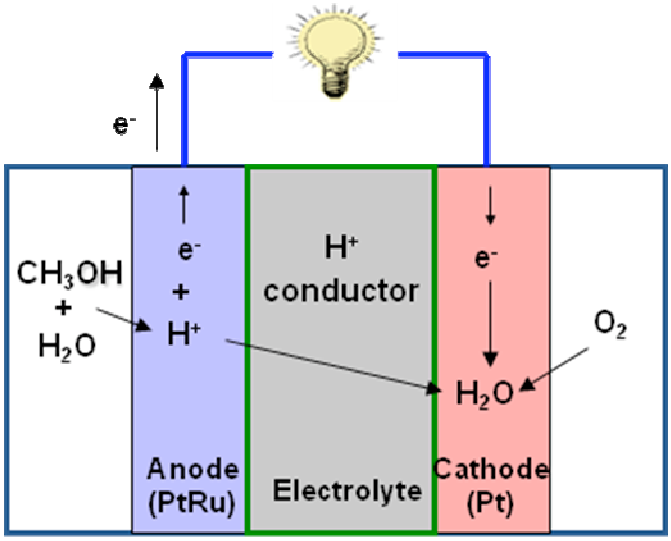

Figure 1: Schematic of a Direct Methanol Fuel Cell (DMFC)

Methanol is supplied to the anode through flow-field channels in the current collector and upon reacting with the catalyst it decomposes into protons (hydrogen ions) and free electrons. The PEM is impermeable to electrons and forces the electrons to travel from the anode to the cathode through an external electrical circuit. These electrons traveling through the external circuit provide the useful energy obtained from the fuel cell. The protons are combined with the externally supplied oxygen or air and the free electrons from the anode to release carbon-dioxide and water (Wilkinson, 2009).

Typically a single DMFC produces a voltage of 0.6 to 0.7 volts only. There are very limited applications which can run on such a low output voltage and hence multiple single cells have to be connected in series to increase the power capacity. A collection of these multiple fuel cells forms a fuel cell stack. To supply fuel (methanol) and oxidant (air/oxygen) to the anode and cathode in successive MEAs, bipolar plates are introduced in between them.

Functions of a bipolar plate

A bipolar plate's primary function is to provide electrical contact between two adjacent MEAs. When the cell is operating, the electrons are taken out from the anode side of the cell and are transferred to the cathode side of the cell to complete the reaction. It is necessary that the direct contact area between the plate and the MEA (called land area) be maximized for easier transfer of electrons from the plate to the MEA (Li, 2004).

The bipolar plate effectively distributes the methanol and air/oxygen to the anode and cathode of the MEA, respectively. This is ensured by introducing a flow-field pattern on the surface of the bipolar plate. These flow-field patterns reduce the land area. The reactants are guided to flow over the active area on either side of the bipolar plate by the flow-field patterns. A proper balance is to be struck between the flow-field pattern's area and the land area to maximize performance of the fuel cell (Li, 2004).

The PEM for DMFC is commonly Nafion. Nafion swells and distorts upon contact with the fuel (Ladewig, 2007), therefore bipolar plates act as a platform to support them. Nafion swelling upon contact with liquid is shown in Figure 2.

Figure 2: Nafion membrane exhibiting distortion and swelling upon contact with liquid

A proper seal is needed between the MEA and the bipolar plate interface to prevent fuel leakage. This is achieved by providing axial force using bolts or some other

fastening mechanism. It is necessary that the bipolar plate needs to withstand this force without cracking or getting crushed. It is also shown that if this axial compressive force is increased, the contact electrical resistance at the MEA-bipolar plate interface is decreased (Cho, 2004).

The electrochemical reactions that occur generate heat and by-products such as water vapor which need to be removed. The bipolar plate needs to be thermally conductive to dissipate heat. The performance of the entire fuel cell system is decreased if this heat is not removed (Hayre, 2006). It is also important to maintain the cell temperature in the operating range by transferring the generated heat efficiently to a heat exchange unit. In some systems, this generated heat is used to heat the reactants to achieve higher efficiency. To eliminate thermal stresses, it is necessary to ensure a low temperature gradient across the plate.

To prevent the methanol and the air/oxygen from mixing in the flow-field, it is important that the bipolar plate is impermeable to the reactant liquid and gases. To reduce the voltage drop due to ohmic losses across the bipolar plate, it should have a high electrical conductivity (Gottesfeld, 1999).

The hot and acidic environment of the fuel cell makes the bipolar plate susceptible to corrosion. Therefore, it should have high corrosion resistance and chemical stability from being oxidized. If an oxide layer is formed at the MEA–bipolar plate interface, it would result in decrease in conductivity and mechanical strength that would ultimately lead to poor fuel cell performance (Gottesfeld, 1999).

State of the art materials for bipolar plates

Currently pure graphite, carbon polymer composites and metals are being used to fabricate bipolar plates. To achieve high corrosion resistance and electrical conductivity, pure graphite has been the preferred material for fuel cell bipolar plates. Graphite

unfortunately has poor machinability and low mechanical strength compared to other materials. These negative properties of graphite make bipolar plates expensive and time consuming to machine. The low mechanical strength of graphite imposes the constraint of the graphite machined plates to be thicker than metal or carbon composite plates, thus adding undesirable weight to the fuel cell.

The benefits of using metallic bipolar plates are reduced plate thickness, higher electrical conductivity and mechanical strength compared to pure graphite bipolar plates. Stainless steel has been used for bipolar plates because of its low cost and superior machinablity when compared to graphite. Unfortunately metals have low corrosion resistance in the DMFC environment and form an electrically non-conductive oxide layer on the surface. Even though the formed oxide layer prevents further corrosion to the bipolar plate, it decreases the electrical conductivity at the MEA–bipolar plate interface. To overcome this pitfall, the metallic plates have to be stamped with the flow-field channel and then coated with thin corrosion resistive surface coating such as gold or titanium nitride (Wang, 2003). These coatings make the metallic bipolar plate expensive (Davies, 2000).

Titanium bipolar plates are also being used due to their lower density, high electrical conductivity and easier formability when compared to stainless steel. Unfortunately, it is too expensive to be used extensively in portable applications where cost is a major factor. Moreover, it also forms titanium oxide on the surface that increases the contact resistance and degrades the fuel cell performance. Similar surface coating strategies could be used to overcome this issue (Hodgson, 2001).

Aluminum is gaining popularity as a material for bipolar plate fabrication because of its light weight and inexpensive fabrication costs. But just like other metals it suffers from high corrosion rates and low chemical stability. Coating gold on aluminum bipolar

plates could reduce the degree of corrosion, but formation of micro-cracks in the surface coating due to mismatch in coefficient of thermal expansion (CTE) degrades the cell's performance (Woodman, 1999).

Multi-phase material systems for bipolar plate fabrication can be classified into carbon- and metal-based. These offer a wide variety of fabrication choices when compared to single-phase material systems (Mehta, 2003).

State of the art bipolar plate manufacturing

The United States Department of Energy (DOE) postulated a series of property requirements for the bipolar plates (Cunningham, 2007). These requirements are tabulated in Table 2.

Table 2: DOE requirements for fuel cell bipolar plates

Electrical conductivity	100 S/cm
Bending strength	25 MPa
Corrosion rate	$< 16\ \mu A/cm^2$
Thermal conductivity	> 10 W/ m K

Graphite has been one of the most preferred materials for the manufacturing of bipolar plates but due to the brittleness of graphite, machining the flow fields becomes expensive and time consuming.

Compression and injection molding are the other alternatives employed to replace the machining process. Meissner *et al* (1998) successfully compression molded bipolar plates using a material system consisting of graphite powder, additives and binders. The bipolar plates obtained from this process required a lot of post processing and do not meet DOE electrical conductivity requirements.

A great improvement in the mass production of compression molded bipolar plates was achieved by the Los Alamos National Laboratory. A mixture of thermosetting vinyl ester resin and graphite powder was used as the material system for this process. To improve strength and electrical conductivity carbon fibers, glass cotton and polymers were included in the material system. This compression-moldable material system has a cure time of less than 30 seconds which revolutionized the mass production of bipolar plates (Wilson, 2001). Unfortunately, the electrical conductivity of these plates did not meet the DOE requirements.

Heinzel *et al* attempted to injection mold bipolar plates using an extruded mixture of carbon compounds and thermoplastic materials in 2004. The cycle time for a single plate was between 30 and 60 seconds. The electrical conductivity of these plates was 50 S/cm and did not meet DOE goals.

Metal foam was used to replace the gas flow fields in bipolar plates by Kumar *et al* (2004). Using metal foam could reduce the weight of the fuel cell stack and simplify stack design by making it act as gas flow field electrode and catalyst support. This material system also suffers from high corrosion like other metallic systems.

NEED FOR THIS RESEARCH

Bipolar plates constitute to 60–80% of the fuel cell stack weight and 40–50% of the total cost of the stack. The complexity and poor performance of the state of the art bipolar plates are major factors in hindering the commercialization of fuel cells. Currently, there is no process that can ensure cost-effective large-scale production of bipolar plates that will meet the DOE targets. It has been established that most promising methods for fabrication of bipolar plates are by injection and compression molding (Scholta, 2004). Compared to other manufacturing processes these two processes are

more beneficial in terms of mass production and cost. Carbon-based composite materials are becoming popular because of their low cost as compared to metals and pure graphite.

Research needs to be done to determine the best material system for the compression or injection molding process to manufacture bipolar plates that meet the DOE requirements. The primary goal of this research is to find the material system and the molding conditions for large-scale production of bipolar plates.

COMPRESSION/INJECTION MOLDING

Compression molding is generally used for thermosetting materials. The material (in powder or granular form) is placed in a heated die where the upper half of the die compresses the material which melts and fills the die cavity. The liquid material fills and conforms to the die shape by the application of pressure. Heat is used to plasticize and then polymerize to make the product permanently hard and hence plays an important role for thermosetting resins. The heat is supplied by steam, heated liquids, electrical resistance or ultra high frequency electric currents. The part solidifies or cures after compression and it is removed by retracting the upper half of the die (Amstead, 1987).

Some thermoplastic materials are processed by compression molding, but the rapid heating and cooling of the mold adds to the difficulty in using such material. The part will be distorted if the mold is not sufficiently cooled before ejection. Mold release agents are generally necessary in molding processes. They prevent the cured part from sticking to the mold and enable easy ejection. Hydraulic presses are commonly used although hand-operated presses are available. Ejector pins are used to force the pins out of the die. The schematic of a general compression molding process is shown in Figure 3 (Amstead, 1987).

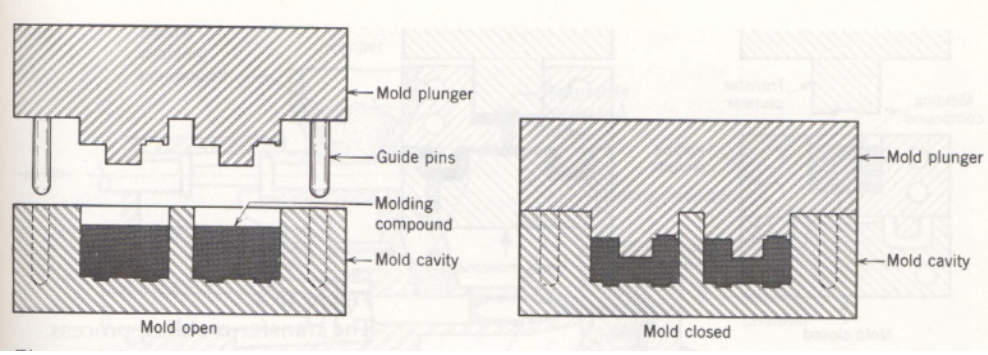

Figure 3: Schematic of compression molding

Injection molding is the process of forcing molten material (usually thermoplastic) under high pressure into a mold cavity. The die must be kept firmly shut during the injection molding process with the aid of mechanical clamps or hydraulic cylinders. The polymer is fed through a hopper to a cylinder or barrel. The die end of this cylinder is surrounded with heaters that gradually bring the polymer to the required operating temperature. Ejector pins are provided for removing the molded components and fine vents ensure that no air remains trapped in the final part. Similar to compression molding, temperature control is very critical in injection molding. Injection molding is preferred where a large number of parts are to be made. A schematic of the injection molding process is shown in Figure 4 (Schey, 1977). A typical injection molding machine is shown in Figure 5.

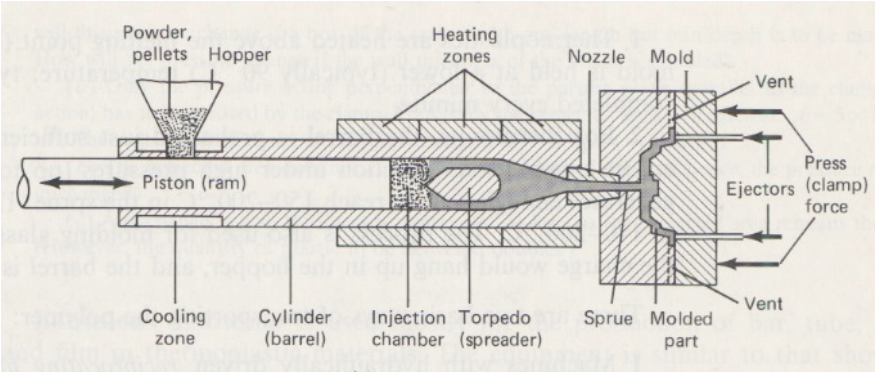

Figure 4: Schematic of a reciprocating piston type injection molding process

Figure 5: An injection molding machine

Reaction injection molding (RIM) differs from injection molding where the reactants (thermoset polymer) are heated and brought together under high pressure inside the mold. Since polymerization takes place inside the mold, the internal stresses are minimal and the process is suitable even for large, complex, filled plastic parts such as auto body and appliance components. RIM is used to scale up the production rate of compression molding.

Chapter 2: *Method of Approach*

Since compression or injection molding is chosen as the preferred manufacturing process, the polymer system should be compatible with the manufacturing process. Secondly, the manufactured bipolar plate must meet the DOE requirements (electrical conductivity, flexural strength and thermal conductivity) for fuel cell bipolar plates and be able to withstand the methanol fuel cell environment.

It is not feasible to mold bipolar plates made out of pure graphite since its melting point is above 3000°C. Hence it is essential to mix graphite with some kind of polymer binder that can be molded.

PERCOLATION THRESHOLD SCIENCE

Electrical and thermal conductivity in a composite can be explained by percolation theory. Percolation theory was originally developed to explain and predict fluid permeation through porous media (Broadbent, 1957). The basis of the percolation model is having a medium with a set of randomly distributed sites into which fluid may flow. The density of these sites determine the probability of formation of interconnecting paths by sites joining together, thereby allowing transfer of fluid through the sample. By analogy, this theory can be used to explain the passage of current and heat through a polymer matrix containing conductive particles. In the composite, the volume fraction of the conductive filler and its uniformity of distribution determine the probability of particles touching and thus forming conductive pathways through the polymeric material.

CANDIDATE MATERIAL SYSTEMS

In this section, several polymers and fillers used in this study will be briefly discussed.

Polymer matrix materials

Epoxy resin

Epoxy is a thermoset resin that cures when mixed with a hardener. Faster curing can be achieved if the curing temperature is increased. For the resin to cure uniformly and exhibit isotropic properties, it is essential that the resin and hardener be mixed thoroughly in the correct ratio. Epoxy is electrically and thermally non-conductive and cannot be used as the bipolar plate material by itself. Hence a composite plate that consists of epoxy as the matrix with electrically conductive filler is a potential solution.

We tested unmodified liquid epoxy resin GY 6010 manufactured by Huntsman Advanced Materials. This liquid epoxy resin is currently used as a coating in the electrical industry, a matrix in the construction industry and a matrix for different composites. It has excellent chemical and heat resistance. It is also compatible with a wide variety of fillers. Aradur 956-2 is a suitable low viscosity liquid amine hardener for GY 6010. The properties of the resin and hardener are shown in Table 3.

Table 3: Property of Araldite GY-6010 and Aradur 956-2

Property	Araldite GY-6010	Aradur 956-2
Viscosity @ 25° C	11000 – 13000 cPs	290 – 500 cPs
Density @ 25° C	1.15 – 1.18 g/cm^3	1.02 g/cm^3
Flash point, closed cup	254°C	> 93°C

Nylon-12

Nylon-12 is a tough thermoplastic and is used in a variety of applications. Since it is a thermoplastic, it can be injection molded easily at temperatures less than 150°C. Nylon-12 is also electrically non-conductive. It exhibits good mechanical properties within its service temperature range (<70°C). Nylon-12 is used as the matrix in

composite materials with reinforcing carbon fibers for added strength and increased density. If an electrically conductive carbon compound is dispersed throughout the matrix such that the percolation threshold is reached, the composite will conduct electricity.

Nylon-12 was obtained in pellet form from Arkema and its properties are listed below in Table 4.

Table 4: Properties of nylon-12

Density	1.03 g/cm^3
Tensile strength	25 MPa
Volume resistivity	11 log ohm.cm
Plasticizing temperature	135°C

Phenolic resin (liquid)

A plasticized phenolic resin (Durite AL 625E) manufactured by Hexion Specialty Chemicals was used in this study. This phenolic resin is ethanol solvated and is highly viscous. It is a thermoset which was designed to be in liquid form to add flexibility for processing. This phenolic resin cross-links at 160°C, making it a good candidate for large-scale production due to its low processing temperature. Properties of Durite AL 625E are shown in Table 5.

Table 5: Properties of Durite AL 625E

Viscosity at 25°C	1400–1800 cPs
Density at 25°C	1.140–1.145 g/cc^3
Solids, by RI	66–73%
Crosslinking temperature	150–160°C
Free Phenol content	7.5–9.5%

Phenolic resin (solid)

We tested a solid phenolic resin in powder form that is easy to process. A Perkin Elmer DSC7 was used to perform Differential Scanning Calorimetry (DSC) analysis on the polymer and the results are shown in Figure 6 (Burns, 1967). The first endothermic peak shows the novolac melting of the resin and the second broad peak represents the heat of solution of hexamine in the molten resin. The exothermic peak around 160°C corresponds to the cross-linking of the novolac and hexamine.

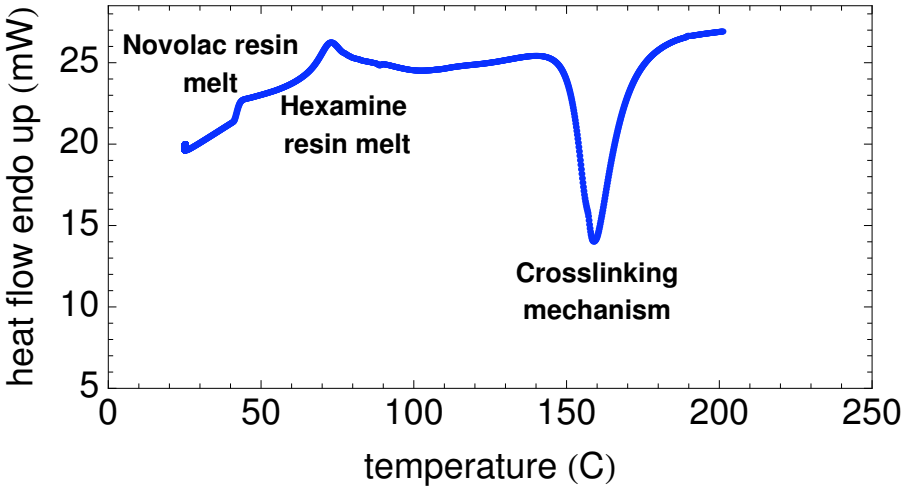

Figure 6: DSC curve of AD 332A phenolic resin

The phenolic resin used in this study was Durite AD 332A, manufactured by Hexion Specialty Chemicals. The particle size distribution is shown in Figure 7. The properties (from datasheet) of the thermoset resin are given in Table 6.

Table 6: Properties of Durite AD 332A

Property	Value
Glass plate flow, 125°C	23–37 mm
Hot plate cure, 150°C	60–90 sec
Hexa content	9.5–10.5%
Particle size	98–100% thru 200 mesh

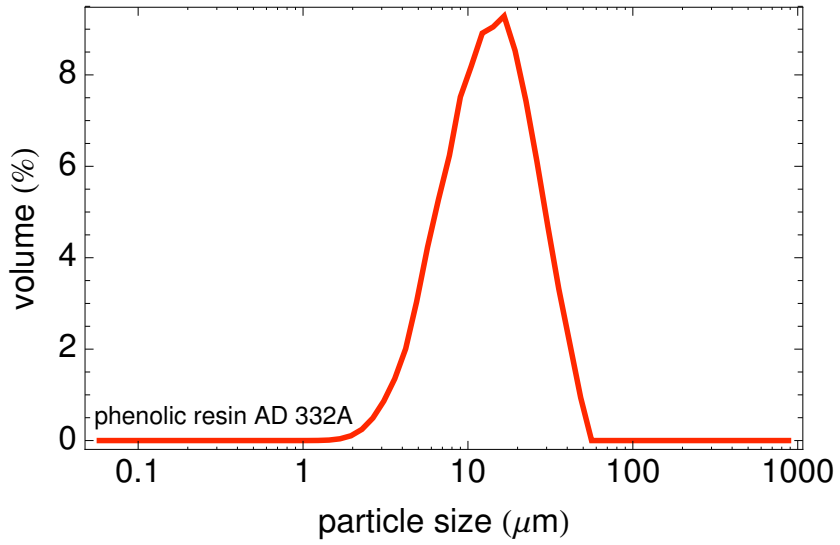

Figure 7: Particle size distribution of AD 332A phenolic resin

Filler materials

Graphite

Graphite is an allotrope of carbon. The carbon atoms form a planar hexagonal arrangement with alternating single and double bonds. These hexagonal layers are held together by weak Van der Waals forces. Graphite is highly electrically conductive and its conductivity is 1.44 x 10^3 S/cm (Kinoshita, 1988). The structure of graphite is shown in

Figure 8. Graphite has to be in powder form in order to be used as filler in our application. It is available in natural and synthetic forms. Graphite is highly chemically inert and thermally stable. Hence it is preferred over metals in a fuel cell.

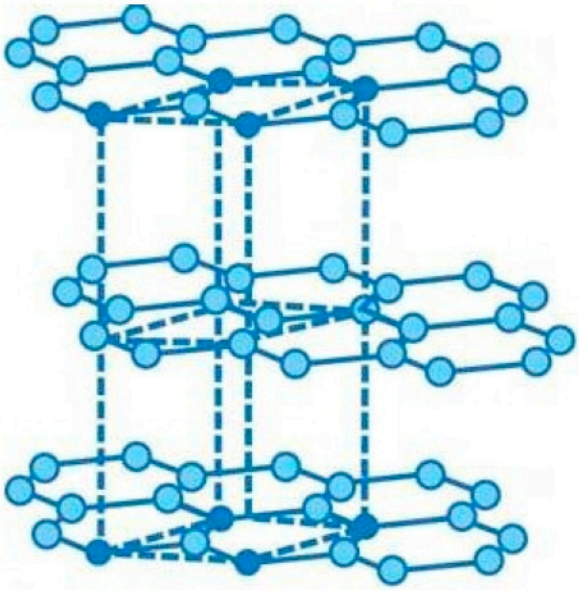

Figure 8: Crystal structure of graphite

Natural graphite

Natural graphite powder is obtained by processing and pulverizing mined graphite. It is available in a wide variety of particle size distributions. The natural graphite used in the experiments was obtained from Alfa Aesar. The particle size was 75 microns (200 mesh). An SEM image showing the particles is shown in Figure 9. A particle size distribution of the natural graphite powder is shown in Figure 10.

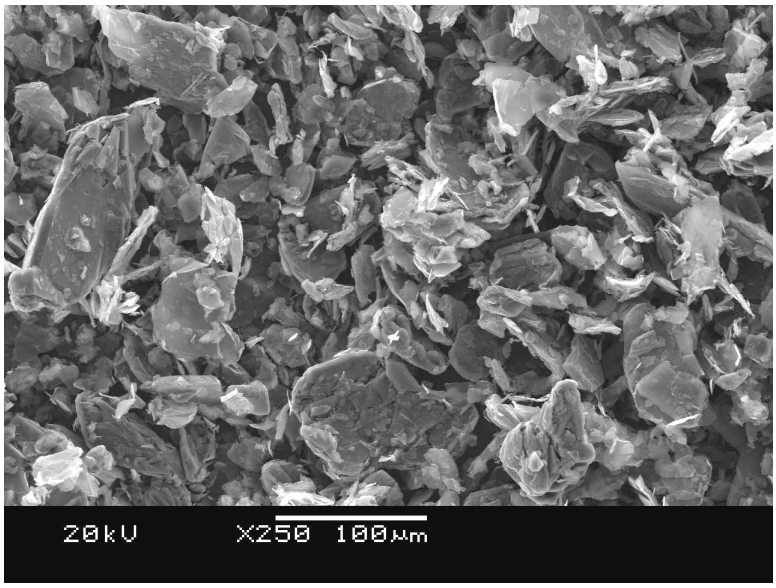

Figure 9: SEM image of 75µm natural graphite powder

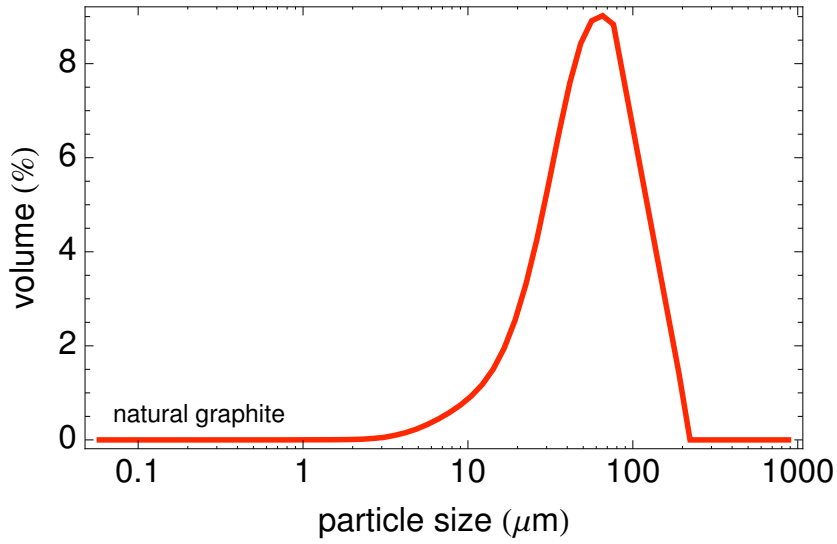

Figure 10: Particle size distribution of natural graphite powder

Synthetic graphite

Synthetic graphite is formed from pitch or coke by graphitization. Graphitization is the process in which amorphous carbon is converted into the hexagonal layers. It

happens around 3000°C (Mantell, 1968). The synthetic graphite we used is called GS150E, supplied by Graftech. The particle size is 75 microns (200 mesh). The particle size distribution is shown in Figure 11. An SEM image showing the particles is shown in Figure 12. Properties of the synthetic and natural graphites are shown in Table 7.

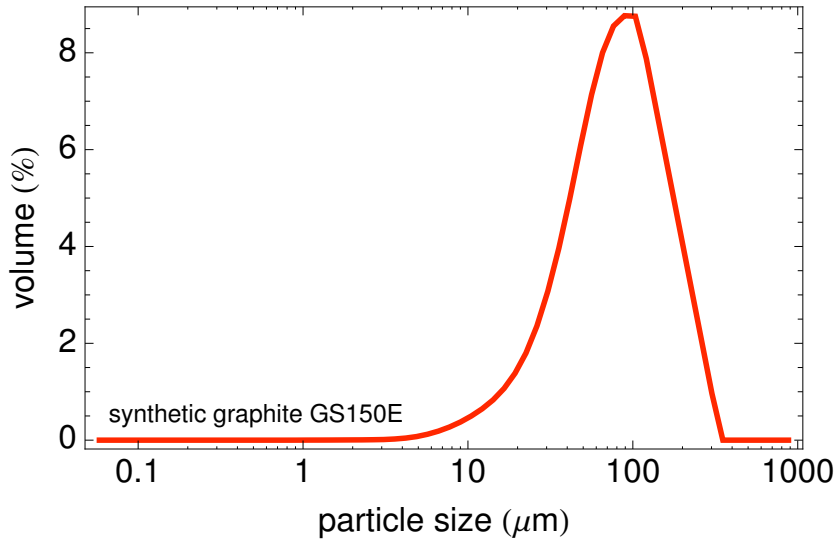

Figure 11: Particle size distribution of GS150E synthetic graphite powder

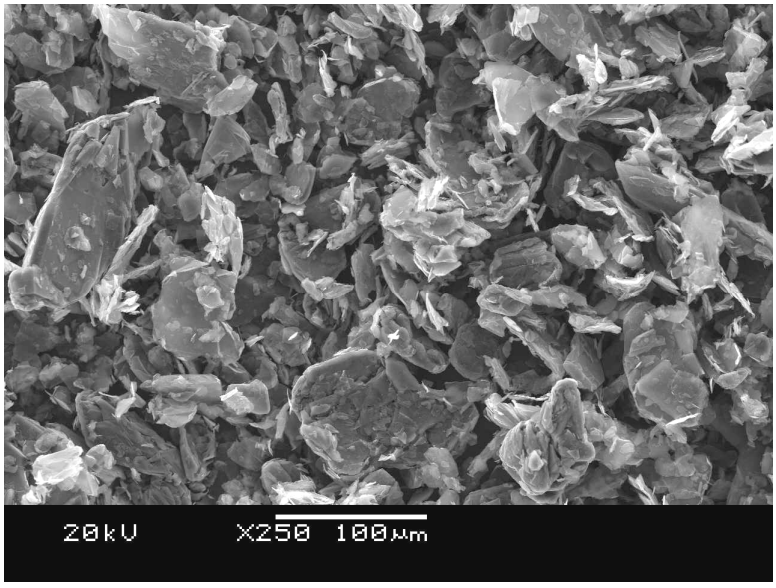

Figure 12: SEM image of GS150E synthetic graphite powder

Table 7: Properties of natural and synthetic graphite

Property	Natural Graphite	Synthetic Graphite
Density	2.25 g/cm^3	2.25 g/cm^3
Cost	-	$4.63/kg
Particle size	75 microns	75 microns

Carbon black

Carbon black is produced by the incomplete combustion of petroleum products. Carbon black is very small (~325 mesh) as a result it has a very high surface area to volume ratio. Like graphite, carbon black is electrically conductive. Because of its high surface area to volume ratio, it is an attractive filler used often in the electrical industry. These properties of carbon black make it a potential candidate filler for fuel cell bipolar plates.

Vulcan XC-72 and XC-605 were obtained from Cabot Corporation. Their properties are listed in Table 8. An SEM image of the carbon black is shown in Figure 13.

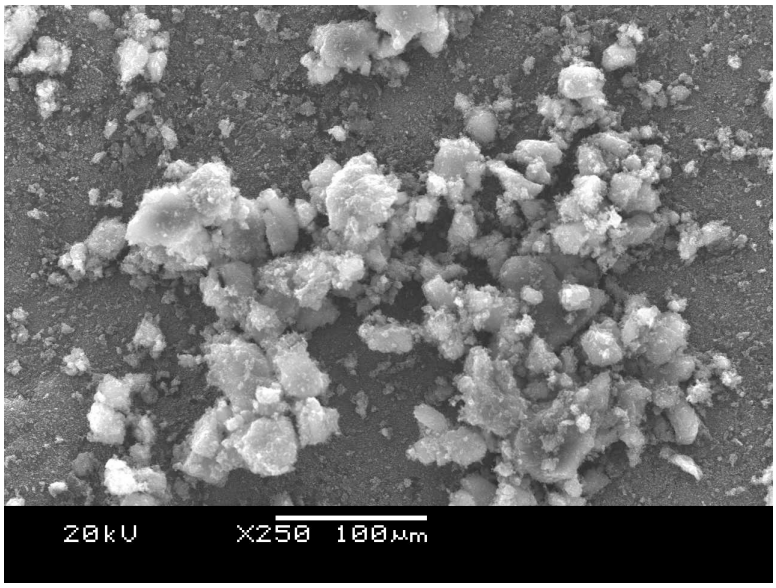

Figure 13: SEM image of Vulcan XC-72 carbon black

Table 8: Properties Vulcan XC-72 and XC-605

	Vulcan XC-72	Vulcan XC-605
Density	2.64 g/cc^3	2.64 g/cc^3
Iodine number	253 mg/g	90 mg/g
325 mesh residue	< 25 ppm	< 20 ppm
Moisture as packed	< 1 %	< 1%

Carbon fiber

Carbon fibers consist of extremely thin fibers composed of carbon atoms. The carbon atoms are bonded and are aligned along the fiber. Carbon fibers posses high strength along the axis and is used often as reinforcements for many materials. Carbon fibers are available as continuous fiber, chopped fiber and woven mats. For dispersion purposes chopped fiber is used (Xanthos, 2005).

T-300 chopped carbon fiber (1/8” long) was obtained from E&L Enterprises. The properties of the carbon fiber are shown in Table 9. An SEM image of the carbon fiber is shown in Figure 14.

Table 9: Properties of T-300 carbon fiber

Density	1.76 gm/cm^3
Specific surface area	0.450 m^2/gm
Electrical resistivity	0.00180 ohm.cm
Thermal conductivity	8.50 W/m.K

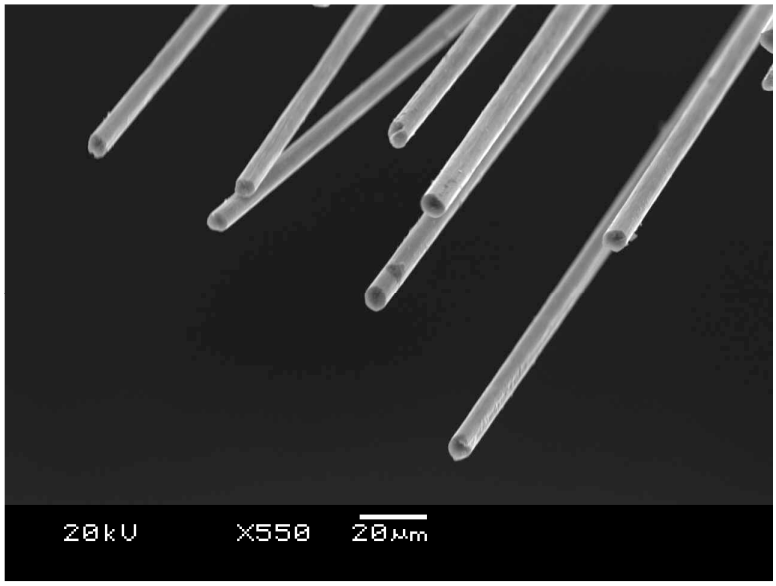

Figure 14: SEM image of carbon fiber

PROCESSING TECHNIQUES FOR CANDIDATE MATERIAL SYSTEMS

Processing of epoxy

Hand mixing

The epoxy resin and hardener were separately mixed with various fillers at different loadings. Because the hand mixing process introduces a lot of air bubbles, the mixture needs to be degassed at 60°C for 12 hours in an oven. Once it was degassed the filled epoxy and hardener were mixed together and cured at 100°C for two hours.

High shear mixer

A high shear mixer usually gives better dispersion of fillers in the resin than the hand mixing technique, especially for highly viscous liquids. The epoxy-carbon filler material system was also mixed using a high shear mixer. The IKA Labor Pilot 2000/4 was used with the MK mixing module. This mixer's minimum mixing capacity is 500 ml. A picture of the machine is shown in Figure 15. The resin was first mixed with the carbon fillers and then the hardener was mixed with the carbon fillers after thorough cleaning to prevent damage to the machine. As the high shear mixing is also a mechanical mixing method, it introduces air bubbles into the mixture, which can be removed by degassing in an oven at 60°C.

The high shear mixing technique can be used only to mix the resin and hardener separately. This machine is incapable of mixing the filled resin and filled hardener with each other as it may solidify in the mixer. Just prior to curing, the filled resin and hardener have to be mixed by hand.

Figure 15: IKA Labor Pilot 2000/4 high shear mixer

Planetary centrifugal mixer

A planetary centrifugal mixer provides uniform stirring by employing rotation and revolution simultaneously. This mixing technique eliminates the need for degassing the mixture after mixing. This mixing technique is very efficient and can completely disperse the particles in less than 30 seconds. A Thinky planetary centrifugal mixer was used and its picture is shown in Figure 16. Similar to the other mixing methods employed for the epoxy-carbon filler system, the epoxy resin and hardener were mixed with the fillers separately and were hand mixed just before curing.

Figure 16: Thinky planetary centrifugal mixer

Processing of Nylon-12

Nylon-12 is a tough polymer and has to be melt-blended. Shear compounding is a technique where two parallel, counter rotating, intermeshing screws create a high shear, high intensity mixing action. It is used to blend, a wide variety of thermoplastics and elastomers. A Brabender shear compounder was used to disperse the filler in the nylon-12. A picture of the shear compounder is shown in Figure 17. Heavy mineral oil was used as the plasticizing agent and the mixing was done at 135°C. The volume of the mixing chamber is 40 cm^3.

The compounded nylon-12 material system takes the form of large cakes when taken out of the compounder. These need to be broken down to a powder form which can be easily handled and compression molded. The pulverization was done in a Wiley mill shown in Figure 18. Photos of nylon-12 pellets, compounded nylon before and after pulverization are shown in Figure 19.

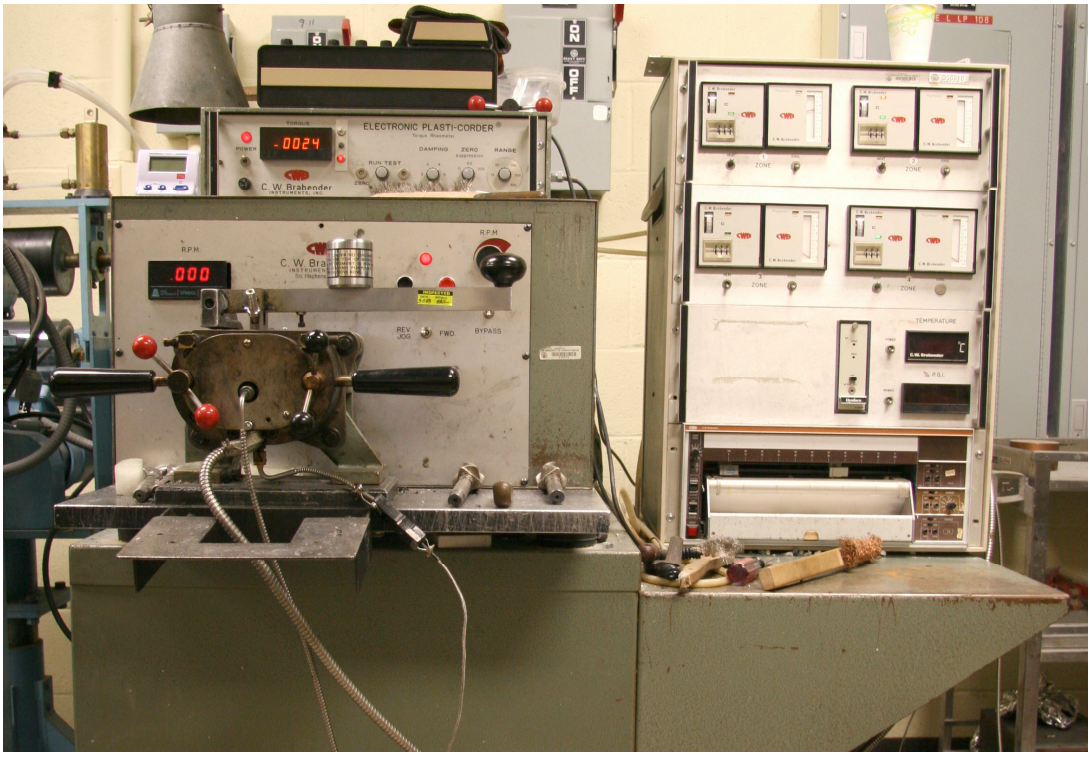

Figure 17: Brabender shear compounder

Figure 18: Wiley mill used to pulverize shear compounded nylon-12 composite

Figure 19: Nylon-12 pellets, shear compounded nylon-12 before and after pulverization

Processing of phenolic resin (liquid)

The viscosity of the liquid phenolic resin is very high. Temperatures as high as 100°C could be reached during the conventional mechanical mixing processes, hence they cannot be used to disperse the carbon fillers in liquid phenolic as the phenolic resin may begin melting. Therefore a non-conventional mixing method is needed.

Since the resin was ethanol solvated, adding ethanol does not change the properties of the resin. Various fillers in different proportions were added to the resin with additional ethanol. This slurry was mechanically mixed at 50°C to evaporate the ethanol. Following evaporation of ethanol, the remaining large agglomerates of powder were pulverized to be an easy precursor for compression/reaction injection molding.

Processing of phenolic resin (solid)

Uniformly mixing two powders is a relatively easy process. A low speed mixing technique is sufficient to mix the carbon fillers and the phenolic. Ball milling was chosen and the powders were mixed in small containers 3" in diameter and 2" high. The grinding media were 5 mm in diameter and were obtained from Glenmills. The grinding media were yttria stabilized high density zirconium oxide.

One third of each jar's volume was filled with the powder, one third with grinding media and one third was left empty. The ball milling was done on a Lortone rock tumbler where the jars spun at 100 rpm. The rock tumbler is shown in Figure 20. After mixing, a sieve was used to separate the powder from the milling media.

Figure 20: Lortone rock tumbler used to ball-mill solid phenolic and carbon filler

Chapter 3: *Evaluation and Selection Process*

PREPARATION OF TEST SPECIMENS

Circular specimens for testing electrical conductivity

Circular discs were molded in a specimen mounting press to test the electrical conductivity of the plates. The specimen mounting press is a quick and easy way to make specimens when compared to compression molding plates. The discs were used to screen each material system by testing if it met the DOE requirements for electrical conductivity.

A Struers LaboPress-3 was used to make the circular discs. A picture of the specimen mounting press is shown in Figure 21. The molding pressure, time and temperature can be adjusted on this machine. A 1.25" diameter ram and cylinder were used. Table 10 shows the parameters used in the LaboPress for molding the specimens.

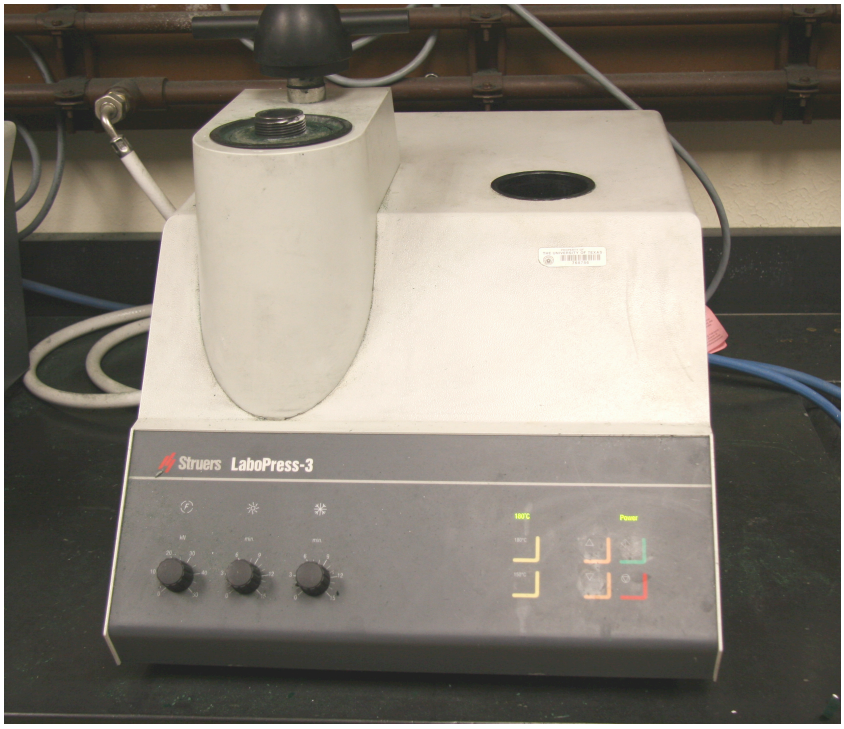

Figure 21: Struers LaboPress-3 used to mold circular disc specimens

Table 10: Molding parameters used on the LaboPress

Temperature	180°C
Pressure	5 – 40 MPa
Heating time	3.5 minutes
Cooling time	3.5 minutes
Mold release	Stoner thermoset mold release
Ram and cylinder arrangement	1.25 inches

Mold release

We tried Stoner thermoset mold release, boron nitride spray and 15% silicone spray for releasing the part from the mold. The commercial mold release agents are shown in Figure 22.

Figure 22: Commercial mold release agents tried

Molding of flat and bipolar plates

Once the material system with electrical conductivity exceeding the DOE target was identified using specimens made with the mounting press, it was necessary to make plates that ensure conformance of other DOE requirements such as flexural strength. Since the tested material systems required a low temperature to cure, aluminum was chosen as the mold material as it is easier to machine and less expensive than tool steel. A punch and die style mold was chosen for versatility in molding plates. With a single die and multiple punches, a wide variety of plate configurations could be molded. The CAD

model of the punch and die style mold is shown in Figure 23. The mold was machined out of Aluminum 6061-T651 and a picture of the mold is shown in Figure 24.

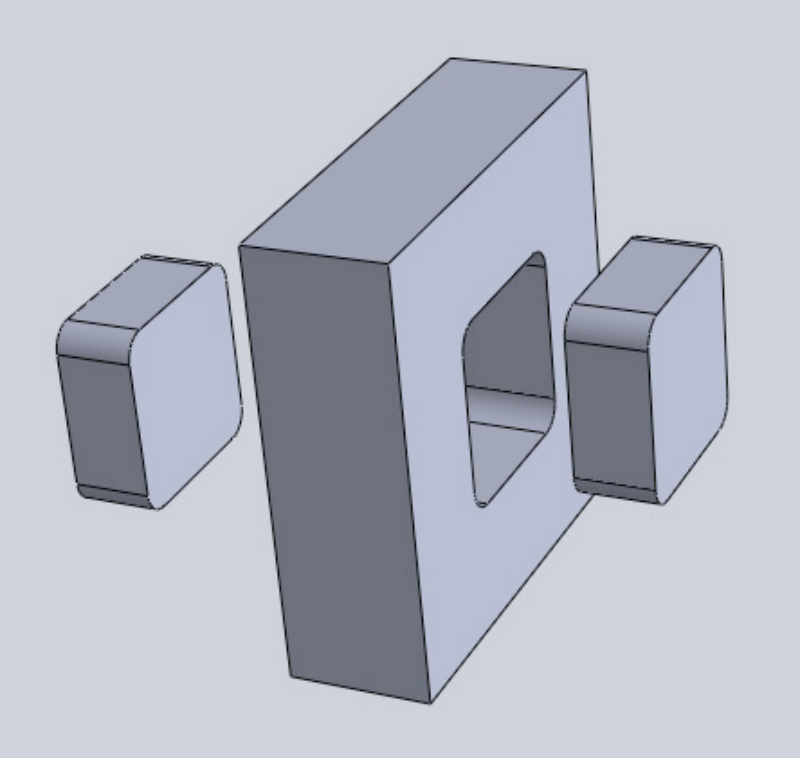

Figure 23: CAD model of punch and die type mold

Figure 24: Punch and die type mold machined from Al 6061-T651

The plates were molded on a Wabash hot press. This machine has two temperature-adjustable heated platens and can mold at various pressures. A picture of the press is shown in Figure 25. Plates with two blank sides (without flow-field channels) were molded to test the electrical conductivity and the flexural strength. To test the performance of the material in a single cell fuel cell, a seven-pass single serpentine bipolar plate was molded. Figure 26 shows the various punches used. Figure 27 shows the CAD model of the flow-field patterns.

Figure 25: Wabash hot press

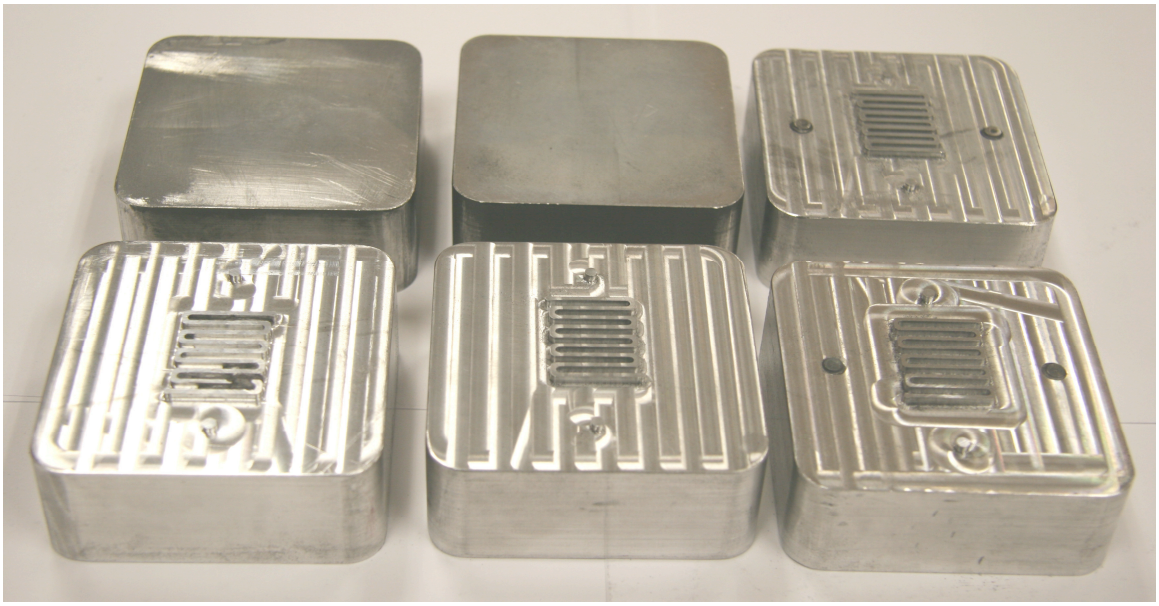

Figure 26: The punches used to mold plates with and without flow field channels

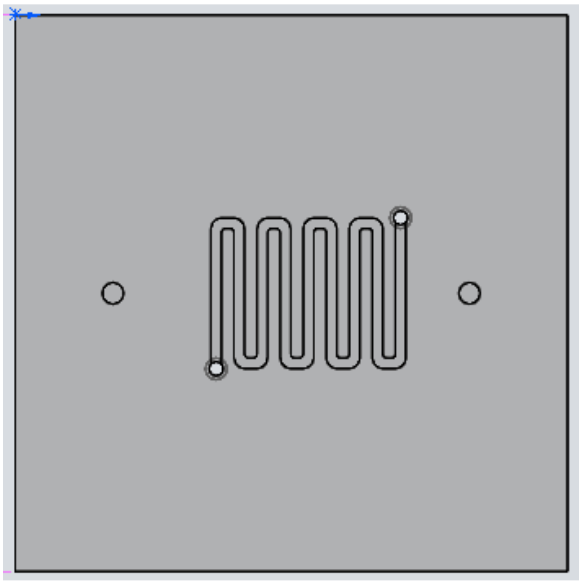

Figure 27: CAD model of the seven-pass single serpentine flow field channel

To make ejection of the part easier, a taper was given to the flow field channels in the mold. The included angle of this taper was two degrees. In addition to the taper, ejection screws were also provided in some punches for easier release of the part.

EVALUATION OF TEST SPECIMENS

Electrical conductivity setup

One of the main requirements of the fuel cell bipolar plate is electrical conductivity. The DOE target is to achieve 100 S/cm of electrical conductivity. Electrical conductivity of highly conductive materials is difficult to measure. Since the potential drop is on the order of 0.5 mV, it requires high-precision instruments.

The four-point probe conductivity measurement technique is used to measure the electrical conductivity of the samples. The experimental schematic of the four point probe technique is shown in Figure 28 (Barbir, 2005).

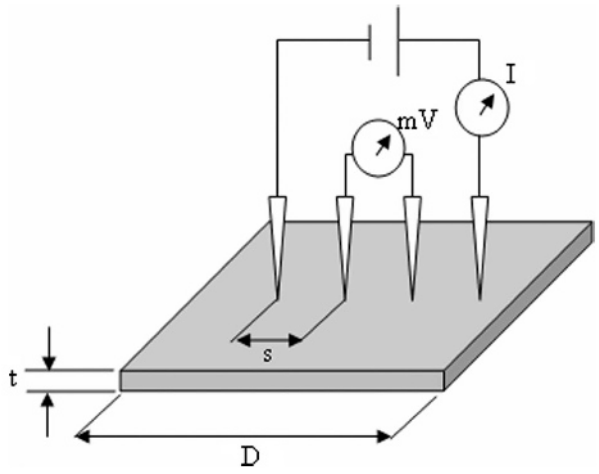

Figure 28: Experimental schematic of four-point probe conductivity setup

The electrical conductivity test was performed using a four-point conductivity probe obtained from Signatone. A picture of the probe is shown in Figure 29. The specifications of the probe are shown in Table 11.

Figure 29: Signatone four-point conductivity probe

Table 11: Specifications of the Signatone four-point conductivity probe

Spacing between tips	1588 microns (62.5 mils)
Spring pressure	85 grams
Tip material	Tungsten carbide
Tip radius	254 microns (10 mils)

The four point conductivity probe was connected to an Agilent power supply and multimeter. The resistivity of the composite plate can be calculated from the following equation (Smits, 1958):

$$C_s = (R_s)^{-1} = [(V / I) \times Cf_1 \times Cf_2]^{-1}$$

where C_s is electrical conductivity, R_s is sheet resistance, V is voltage drop, I is current through the probes, Cf_1 is the correction factor for the D/s ratio, Cf_2 is the correction factor for t/s ratio, s is spacing between probe tips and t is sample thickness.

Five specimens were prepared for each blend and 20 measurements were taken on each specimen.

Three-point bending test setup

The three point bending test was performed on an Instron 3345 in accordance with the ASTM D 790-07 test standard. This standard is used to determine the flexural properties of unreinforced and reinforced plastics, including high-modulus composites and electrically insulating materials in the form of rectangular bars molded directly or cut from sheets. The specimen dimensions were 76 by 12.7 by 3.2 mm. Square plates were molded without flow channels and were sanded down to the required dimension on a belt sander. A picture of the three-point bending test setup is shown in Figure 30.

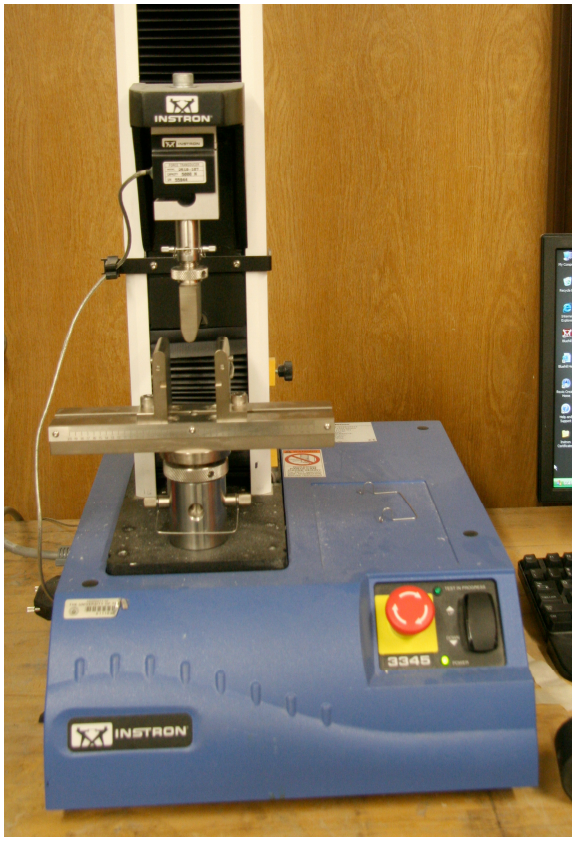

Figure 30: Instron 3345 set up for the 3 point bending test with 50 mm jaw spacing

Thermal conductivity setup

A steady-state longitudinal heat flow method was used to determine the thermal conductivity of the composites. The principle of this method is that when heat flux is passed through a sample with known thermal conductivity and through an unknown sample, the thermal gradients are inversely proportional to their thermal conductivities. Thermal conductivity can be obtained from the following equation:

$$\kappa = -\frac{q\Delta x_s}{A_s \Delta T_s} = -(\frac{\kappa_r . A_r . \Delta T_r}{\Delta x_r}).\frac{\Delta x_s}{A_s . \Delta T_s}$$

where, κ is thermal conductivity, q is the heat flow, Δx is the distance between thermocouples on reference (r) or sample (s), ΔT is the temperature difference along the heat flowing direction across a distance Δx on reference (r) or sample (s), A is the cross sectional area of reference (r) or sample (s).

EVALUATION AND SELECTION

Epoxy

Hand mixing

Filler loading between 10% and 90% (by volume) were tested. None of these blends was electrically conductive. Since these were mixed by hand, the mixing was non-uniform and trapped air bubbles in the final resin-hardener mixture. It was concluded that hand mixing epoxy and filler for a bipolar plate was not a feasible production process.

IKA high shear mixer

The IKA high shear mixer was unable to mix at filler concentrations above 40% by volume. This was because the viscosity of the mixture exceeded the machine's limit. It

was difficult to hand mix the filled resin and filled hardener. It resulted in improper mixing and parts with poor surface finish which were electrically non-conductive.

Thinky planetary mixer

Attempts were made to increase the filler concentration in the resin and the hardener. With the Thinky mixer we were able to increase the loading up 75% by volume, but the viscosity of the mixture was so high that it did not flow out of the jar. We found it impossible to blend the resin and hardener. Experiments performed with epoxy and hardener did not yield any useful results in the manufacture of the bipolar plate.

Nylon-12

Various fillers were tried with nylon-12. About 50% by volume was the maximum loading which was reached with different fillers. The shear compounder was not able to compound above this loading level. The fillers used and their conductivities are shown in Table 12. Some carbon fiber was added to improve strength, but the shear compounder was not able to disperse the fibers and they were in large clumps.

Table 12: Fillers used and maximum electrical conductivity obtained with nylon-12

Filler	Maximum conductivity at maximum attainable loading
Synthetic graphite	2 S/cm
Carbon black	12 S/cm
Natural graphite	10 S/cm
Natural graphite + carbon black hybrid	34 S/cm

The electrical conductivity of the nylon-12 composite was much lower than the DOE goal. A possible reason for this could be that the shear compounder was unable to disperse the filler uniformly as nylon-12 is a tough matrix.

Phenolic resin (liquid)

The composition shown in Table 13 yielded the best conductivity (136 S/cm). This conductivity was higher than the DOE target. The composite with phenolic as the matrix showed good potential but a better and more effective method for mixing was needed. Mixing in excess alcohol is expensive and is a process that cannot be scaled up. Moreover, releasing alcohol into the atmosphere is damaging to the environment.

Table 13: Composition for the composite with best electrical conductivity for liquid phenolic resin matrix

Component	Concentration in vol%
Liquid phenolic resin	48%
Natural graphite	37%
Carbon black	15%

Phenolic resin (solid)

The electrical conductivity of the composite plate depends on the graphite content, molding pressure, and mixing time on the ball mill.

Effect of graphite content

Electrical conductivity was found to be directly proportional to the graphite content. This is because graphite is the electrically conductive component of the mixture; hence increase of graphite should increase the overall conductivity of the bipolar plate. The DOE target of 100 S/cm was reached with graphite filler concentration of 80% by volume. When the graphite content was increased to 87.5% by volume, the electrical conductivity was 160 S/cm. Figure 31 shows the electrical conductivity vs. graphite content for plates molded at 12 MPa. The shaded pink band around the conductivity shows the 95% confidence interval.

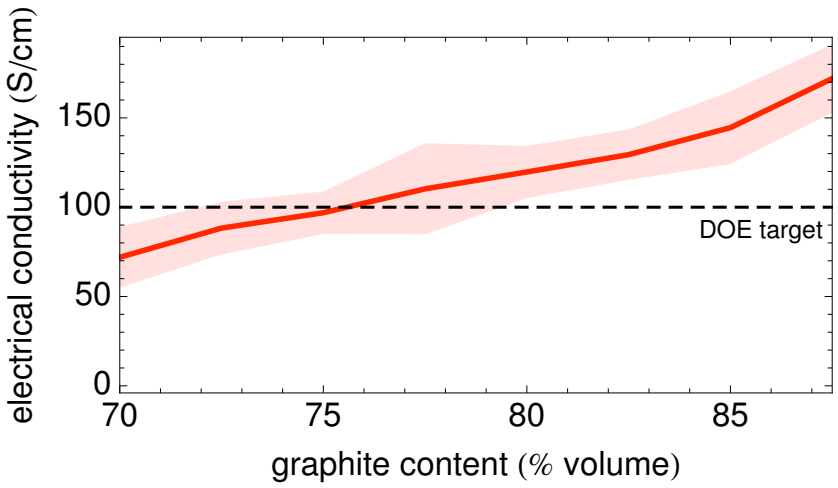

Figure 31: Electrical conductivity vs. synthetic graphite concentration (molded at 12 MPa)

Effect of molding pressure

It was found that the molding pressure has a significant effect on electrical conductivity until 15 MPa after which increase in pressure does not affect electrical conductivity. Figure 32 shows the electrical conductivity vs. molding pressure for 87.5% by volume concentration of graphite. Increase in molding pressure results in tighter compaction of the moldable mixture. Hence the electrically conductive graphite particles are brought closer together enabling higher electrical conductivity.

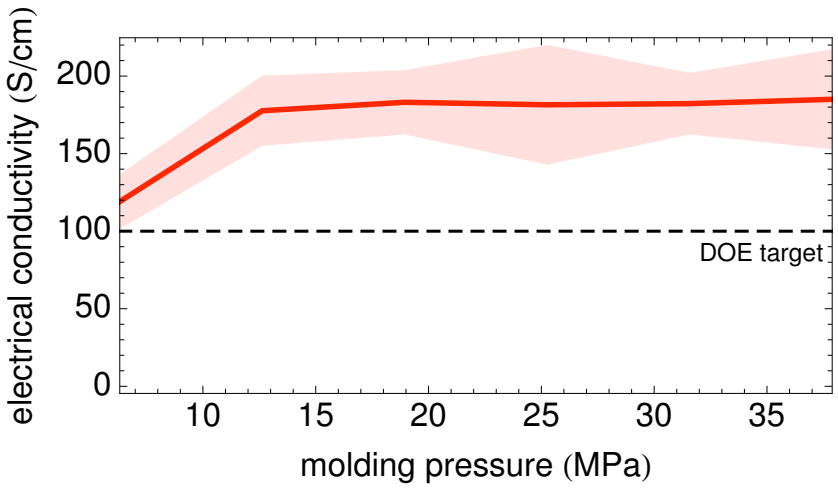

Figure 32: Electrical conductivity vs molding pressure (87.5% by volume concentration)

The blend with 87.5% by volume synthetic graphite and 12.5% by volume solid phenolic powder was found to have the best electrical conductivity. This was chosen as the optimal composition to mold fuel cell bipolar plates.

Effect of processing time on the ball mill

The processing technique also seemed to have an influence on the electrical conductivity. The electrical conductivity was found to be inversely proportional to the mixing time on the ball mill. Figure 33 shows the electrical conductivity vs. mixing time on the ball mill for two different molding pressures. Mixing for three hours on the ball mill is sufficient to yield plates of good electrical conductivity. This could be because mixing for longer alters the particle size distribution by making it smaller and it is difficult for the particles to touch each other and conduct electricity.

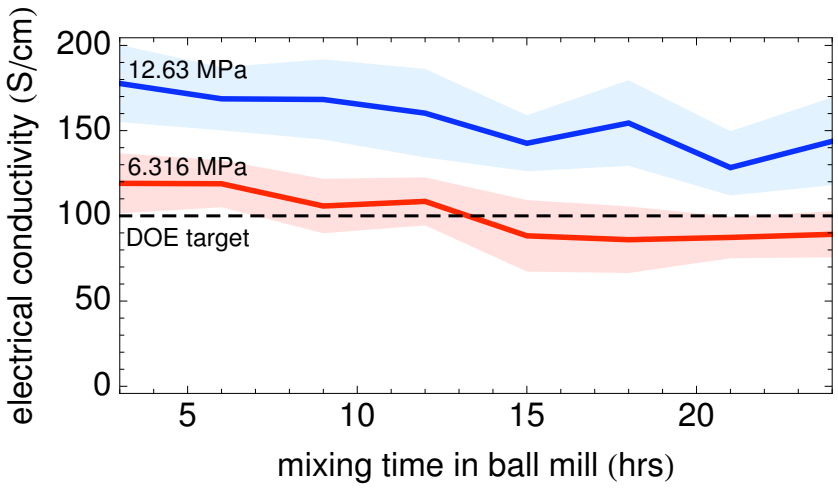

Figure 33: Electrical conductivity vs mixing time in ball mill (87.5% by volume concentration)

Flexural strength

The flexural strength of the composite molded from the optimal composition was found to be 40 MPa. This is much higher than the DOE requirement which is 25 MPa.

Thermal conductivity

The thermal conductivity of the molded from the optimal composition was found to be 87.5 W/m K. This is much higher than the DOE requirement which is 10 W/m K.

Chapter 4: *Single-cell Fuel Cell Tests*

The performance of the molded bipolar plates was evaluated in an 890e Multi-range fuel cell testing system bought from Scribner Associates Inc, shown in Figure 34. In this commercial fuel cell test system, hardware for a single cell fuel cell bought from Fuel Cell Technologies was installed. The 5 cm^2 molded plates were compared against a 5 cm^2 active area, 11 pass single serpentine plates. These commercial plates were machined from natural graphite.

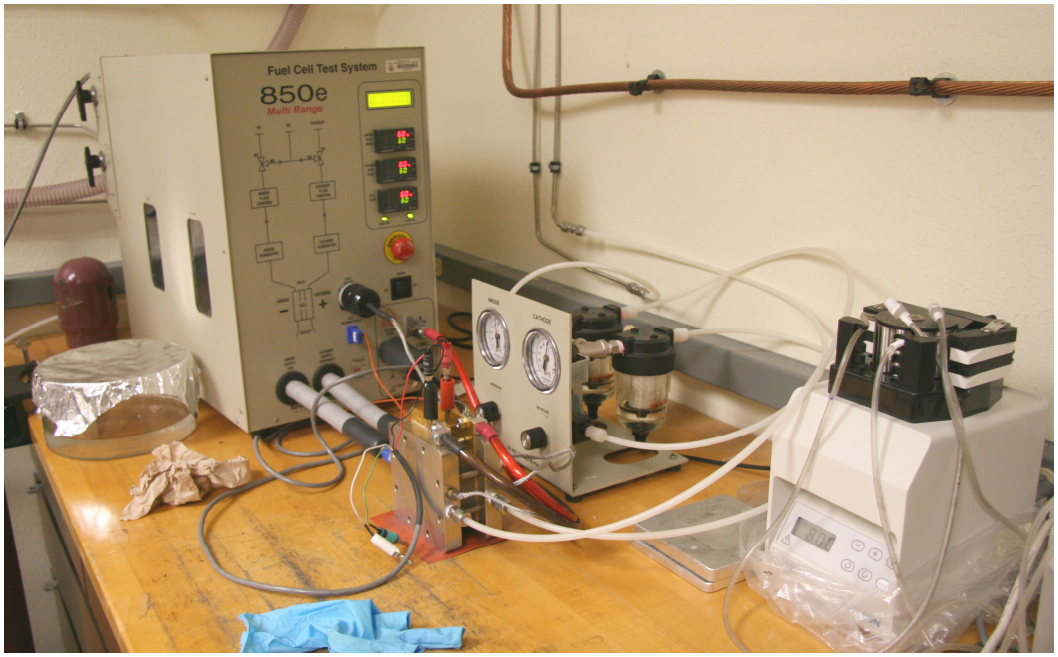

Figure 34: 890e Multi-range fuel cell testing system

MEMBRANE ELECTRODE ASSEMBLY PREPARATION

The electrodes consisting of gas-diffusion and catalyst layers were prepared as reported in (Li, 2008). The anode catalyst consisted of 60 wt% Pt-Ru (1:1) on Vulcan carbon (obtained from E-TEK). The cathode catalyst was 60 wt% Pt on Vulcan carbon

(obtained from Alfa Aesar). The prepared electrodes were impregnated with a 5 wt% Nafion solution (obtained from DuPont Fluoroproducts) by spraying. It was then dried at 90°C under vacuum for 30 minutes. The Nafion-115 loading for the anode and cathode catalysts were 0.35 mg/cm^2. A Carver Inc. Model 3851-0 was used to hot press the membrane, anode and cathode together. The anode, membrane and the cathode were arranged between two sheets of aluminum foil and pressed at 140°C for 3 minutes.

SINGLE CELL TEST METHOD

A 1M methanol solution was stored and heated in a glass flask using a heating mantle bought from Electrochemical Engineering Ltd. The temperature of the methanol was controlled and was set to match the fuel cell's operating temperature which was 65°C. The glass flask has four ports. One port is to supplies methanol to the pump and another port collects methanol back from the cell. The other two ports are for a temperature probe and an air condenser. Preheated methanol was fed into the anode side of the cell at the rate of 2.5 ml/min controlled by a peristaltic pump without back pressure. Oxygen was fed to the cathode side at 200 ml/min without back pressure. The humidifier's temperature was set at 65°C to match the cell's operating temperature.

METHANOL CROSSOVER EVALUATION

A voltammetric method (Ren, 2000) was used to determine the methanol crossover. 1M methanol solution was fed into the anode side of the cell when the cathode side was supplied humidified N_2 gas. A positive potential was applied to the cathode side and the steady-state limiting current density from the complete electro-oxidation at the cathode side Nafion/Pt catalyst interface was used to determine the flux rate of the permeating methanol.

SINGLE CELL TEST RESULTS

The polarization curve obtained from the single cell fuel cell that used the molded bipolar plate is shown in Figure 35. It is compared against a polarization curve from a commercially machined graphite plate single cell fuel cell as shown in Figure 36.

The open circuit potential (OCV) can be obtained from the initial part of the polarization curve (near zero current density). The higher the polarization curve, the lower is the methanol crossover. In the commercial plate and the molded bipolar plate the open circuit voltage is about 0.65 volts, demonstrates that both the plates have very similar methanol crossover characteristics.

The general operating potential of a cell is 0.4 volts and the current density at this potential determines the power output of the cell. The commercial plate shows a marginally superior current density at this potential, but this could be due to the different flow-field patterns. The primary function of the flow-field pattern is to distribute the reactants over the MEA, hence it plays a very important role in the performance of the cell.

The maximum current density of the fuel cell is at around 0.2 volts. The commercially machined graphite bipolar plate has a current density of 245 mA/cm^2 when compared to the molded plate's 210 mA/cm^2 at 0.2 volts. Since the fuel cells were identical in both the tests except for the bipolar plates, it shows that the commercial plate's electrical resistance is lower than the molded plates. This is expected as the commercial plate is made from pure graphite (electrical conductivity is over a 1000 S/cm) when the molded plate's conductivity is 160 S/cm. The commercially machined bipolar plate is definitely more electrically conductive than the molded plate but cannot be produced in large quantities effectively. The molded bipolar plate can be produced in

large quantities effectively and also meets the DOE requirements. Hence, there is a tradeoff between loss in performance and the ability to be produced in large quantities.

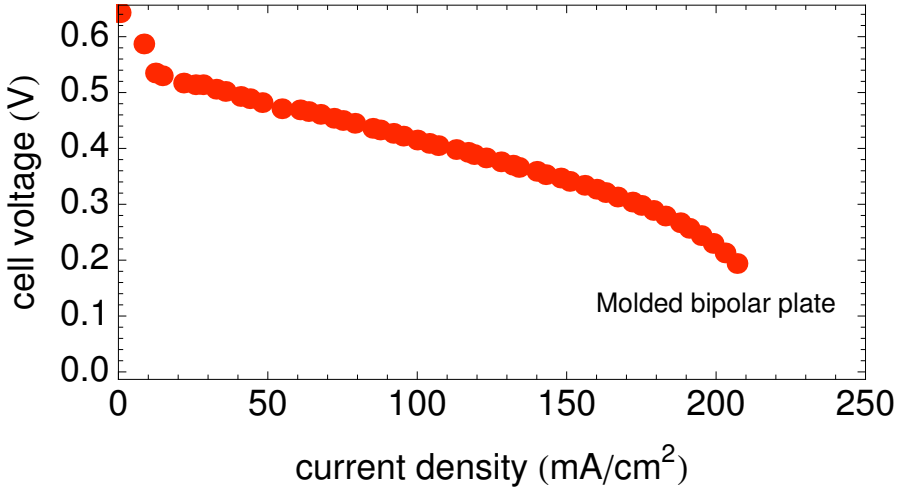

Figure 35: Polarization curve for a 5 cm^2, seven-pass single serpentine molded bipolar plate

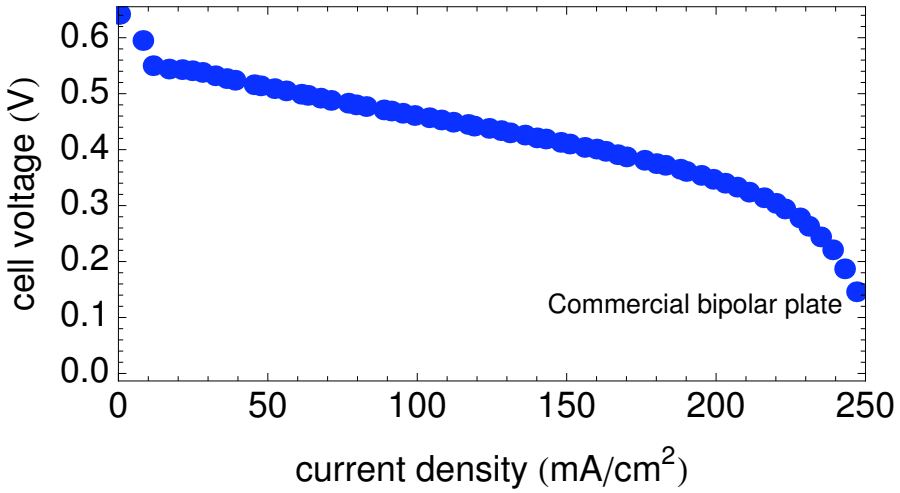

Figure 36: Polarization curve for a 5 cm^2, eleven-pass single serpentine commercial bipolar plate

METHANOL CROSSOVER TEST RESULTS

The methanol crossover test results obtained for the molded and commercial bipolar plates are shown in Figures 37 and 38 respectively. The steady state current density (usually between 900-1000 mV) is a measure of methanol crossover. The molded and the commercial plates exhibit very similar methanol crossover characteristics.

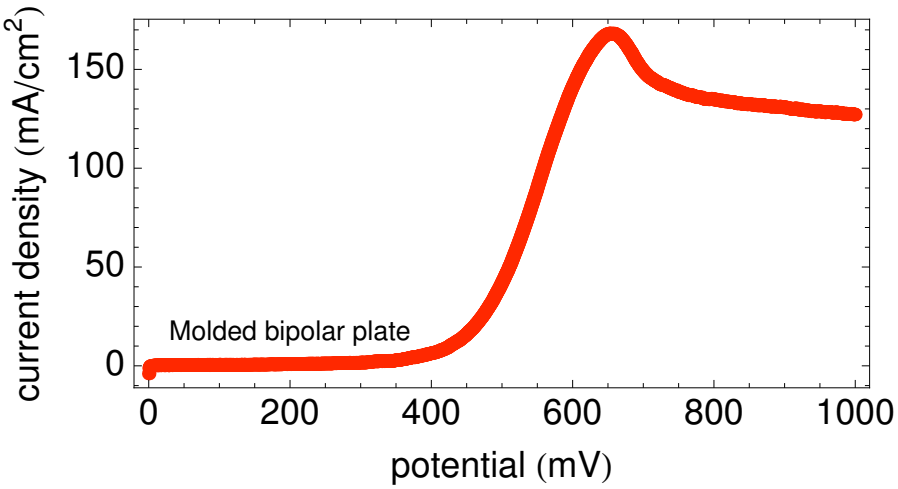

Figure 37: Methanol crossover for the molded bipolar plates

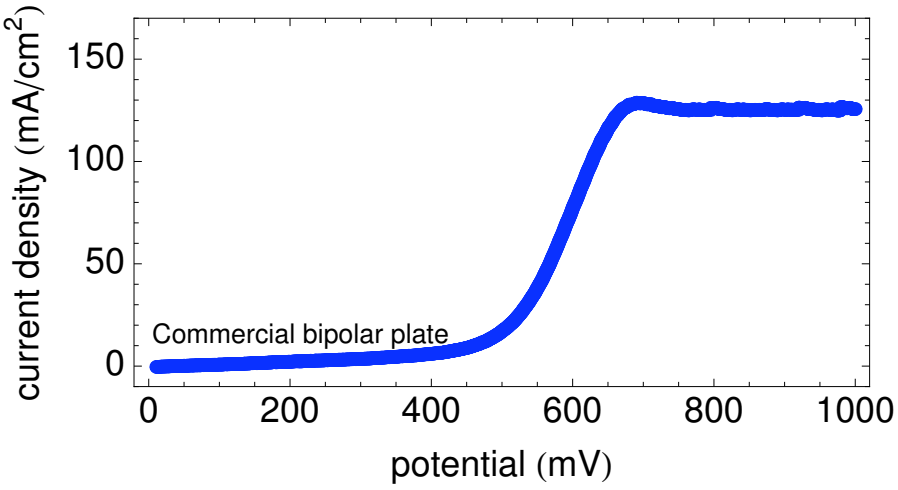

Figure 38: Methanol crossover for the commercial bipolar plates

Chapter 5: *Summary, Conclusion and Future Work*

SUMMARY AND CONCLUSION

Fuel cells are promising energy conversion devices. The high manufacturing cost of bipolar plates is one of the reasons that prevent commercialization of fuel cells. Based on literature review, it was found that compression or injection molding is a suitable process for large-scale manufacturing of bipolar plates. This research aims at defining materials and molding parameters for the same process.

Epoxy resin, nylon-12, liquid phenolic resin and solid phenolic resin were the candidate matrix materials tested. Natural and synthetic graphite powder, carbon black and carbon fibers were the fillers tested. It was concluded that epoxy resin is an unsuitable matrix due to its high viscosity and the related mixing difficulties. Due to the high toughness of nylon-12, the dispersion of fillers is very difficult and hence it is not suitable either. Composites with liquid phenolic resin as the matrix exhibited electrical conductivity that exceeded the DOE requirements. Unfortunately this process is not scalable since the preparation of the mixture involves mechanical mixing in excess of alcohol.

The composites with solid phenolic resin as the matrix and synthetic graphite as the filler surpassed all the DOE requirements. Best properties were obtained when the powder was mixed in a ball mill for three hours. The composition, molding conditions and properties of the best mixture are shown in Table 14. Single serpentine seven-pass bipolar plates were molded its performance was compared against a commercially available bipolar plate. A picture of the molded bipolar plates is shown in Figure 39.

Table 14: Composition, molding conditions & properties of the optimal mixture

Synthetic graphite content	87.5% by volume
Solid phenolic content	12.5% by volume
Processing time in ball mill	3 hours
Molding temperature	165°C
Molding pressure	23 MPa
Mold release agent	Stoner thermoset mold release
Mold material	Aluminum
Electrical conductivity	160 S/cm
Thermal conductivity	87.5 W/mK
Flexural strength	40 MPa
Density	1.95 g/cm^3

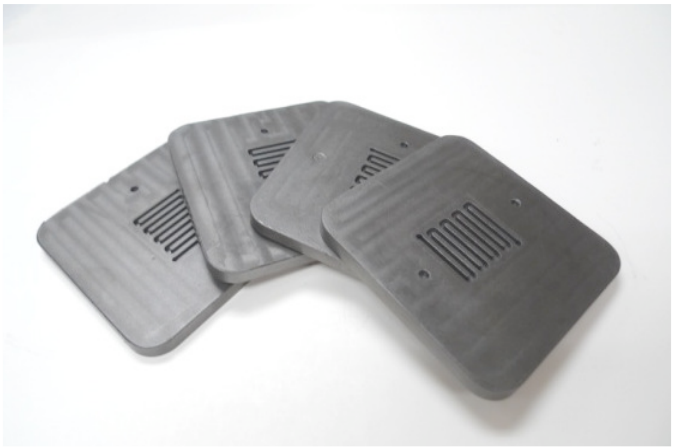

Figure 39: Plates molded from solid phenolic resin and synthetic graphite

FUTURE WORK

The identified optimal composition as prepared in small batches in the laboratory. Work needs to be done in defining and scaling up the processing of the graphite and

phenolic. Even though the molding conditions were identified for the optimal composition, it is necessary to properly quantify the molding parameters for a high-volume injection molding process.

Even though the cost of manufacturing the mold is greatly reduced by using aluminum, it might be slow to machine a mold with complex flow field geometries. It might be better to rapidly prototype the mold using Selective Laser Sintering (SLS).

The long term behavior of this molded fuel cell bipolar plate was not analyzed. Study needs to be done to quantify interaction with methanol to see if there is any significant drop in electrical conductivity or if there is something leaching out of the plates into the solution.

The flow-field channels on the fuel cell bipolar plate dictates the interaction of the fuel with the MEA. It plays a critical role in the performance of the fuel cell and it needs to be optimized. A Computational Fluid Dynamics (CFD) model to simulate the bipolar plate-MEA interface needs to be built and the flow-field channels need to be optimized for maximum performance.

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Vita

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